

## Supplementary Information

### Designing Mixed Metal Halide Ammines for Ammonia Storage Using Density Functional Theory and Genetic Algorithms

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## Starting Populations

As explained in the methods section, the starting populations are chosen randomly with compositions fulfilling the criteria, defined in the methods section. The actual chosen starting structures are:

1st run		2nd run		3rd run	
Composition	wH	Composition	wH	Composition	wH
Ni <sub>2</sub> FeCdCl <sub>8</sub>	7.42%	Cu <sub>2</sub> CoBeBr <sub>4</sub> Cl <sub>4</sub>	6.81%	FeCo <sub>2</sub> BeBr <sub>4</sub> Cl <sub>4</sub>	6.89%
ZnMn <sub>2</sub> CrBr <sub>4</sub> Cl <sub>4</sub>	6.61%	V <sub>2</sub> MnFeBr <sub>4</sub> Cl <sub>4</sub>	6.70%	SrNbCr <sub>2</sub> Br <sub>4</sub> Cl <sub>4</sub>	6.29%
ZnNi <sub>2</sub> NbBr <sub>4</sub> Cl <sub>4</sub>	6.33%	ScMn <sub>2</sub> CuBr <sub>4</sub> Cl <sub>4</sub>	6.67%	YMo <sub>2</sub> MgBr <sub>4</sub> Cl <sub>4</sub>	6.18%
ZrRhBa <sub>2</sub> Cl <sub>8</sub>	6.25%	Ni <sub>2</sub> MoCaBr <sub>4</sub> Cl <sub>4</sub>	6.46%	MoMg <sub>2</sub> CuCl <sub>4</sub> I <sub>4</sub>	5.73%
Zr <sub>2</sub> TiRuBr <sub>4</sub> Cl <sub>4</sub>	6.04%	Ti <sub>2</sub> SrPdBr <sub>4</sub> Cl <sub>4</sub>	6.26%	YCoCa <sub>2</sub> Cl <sub>4</sub> I <sub>4</sub>	5.64%
PdFe <sub>2</sub> BaBr <sub>4</sub> Cl <sub>4</sub>	5.92%	ZrCr <sub>2</sub> BaBr <sub>4</sub> Cl <sub>4</sub>	6.03%	Y <sub>2</sub> CuBeCl <sub>4</sub> I <sub>4</sub>	5.55%
CuCoBa <sub>2</sub> Br <sub>4</sub> Cl <sub>4</sub>	5.73%	Zr <sub>2</sub> FeBaBr <sub>4</sub> Cl <sub>4</sub>	5.83%	SrPdBa <sub>2</sub> Br <sub>4</sub> Cl <sub>4</sub>	5.42%
Y <sub>2</sub> NbMgBr <sub>8</sub>	5.40%	ZrNi <sub>2</sub> BeBr <sub>8</sub>	5.73%	VSr <sub>2</sub> NiCl <sub>4</sub> I <sub>4</sub>	5.40%
Zr <sub>2</sub> NiMoCl <sub>4</sub> I <sub>4</sub>	5.20%	Cu <sub>3</sub> CrCl <sub>4</sub> I <sub>4</sub>	5.58%	YCa <sub>2</sub> BaCl <sub>4</sub> I <sub>4</sub>	5.32%
Y <sub>2</sub> CuAgBr <sub>8</sub>	5.19%	RuNi <sub>2</sub> CaCl <sub>4</sub> I <sub>4</sub>	5.51%	ZrYC <sub>2</sub> Cl <sub>4</sub> I <sub>4</sub>	5.32%
Y <sub>3</sub> RuBr <sub>8</sub>	5.13%	RhNiCr <sub>2</sub> Cl <sub>4</sub> I <sub>4</sub>	5.48%	Zn <sub>2</sub> VTiBr <sub>4</sub> I <sub>4</sub>	4.95%
Zn <sub>2</sub> PdBaCl <sub>4</sub> I <sub>4</sub>	5.07%	Mn <sub>2</sub> FeCdCl <sub>4</sub> I <sub>4</sub>	5.43%	ScCdCa <sub>2</sub> Br <sub>4</sub> I <sub>4</sub>	4.93%
SrPdMn <sub>2</sub> Br <sub>4</sub> I <sub>4</sub>	4.71%	Zr <sub>2</sub> BeBaCl <sub>4</sub> I <sub>4</sub>	5.23%	MgCu <sub>2</sub> BaBr <sub>4</sub> I <sub>4</sub>	4.76%
Mg <sub>2</sub> CoAgI <sub>8</sub>	4.43%	Sr <sub>2</sub> NbBaBr <sub>4</sub> I <sub>4</sub>	4.42%	Ca <sub>2</sub> BaAgBr <sub>4</sub> I <sub>4</sub>	4.65%
ZnSr <sub>2</sub> MgI <sub>8</sub>	4.30%	ZrNbCo <sub>2</sub> I <sub>8</sub>	4.21%	SrMo <sub>2</sub> CuBr <sub>4</sub> I <sub>4</sub>	4.60%

Table S1: Starting structures for the three test runs.

## Top 10 Candidates Detailed

1st run:

Composition	Fitness	$\Delta H_{60}$	E_decomp_0	E_decomp_1	E_decomp_2	E_decomp_6	$\Delta H_{60}-\Delta H_{61}$	$\Delta H_{60}-\Delta H_{62}$	wH pract.
TiScTiCaCl8	8.31%	54.8	22.0	14.1	11.1	25.3	0.9	-3.9	8.31%
TiTiCuMgCl8	8.28%	54.9	-34.0	-20.0	-40.2	-19.2	-0.9	5.2	5.52%
TiTiTiScCl8	8.24%	53.0	0.9	0.1	-1.4	-0.2	-0.7	-5.2	8.24%
TiScTiVCl8	8.21%	54.1	-2.4	-1.6	-5.8	-5.0	-1.1	-4.4	8.21%
TiScTiCrCl8	8.20%	52.6	-8.5	-3.4	-13.8	7.7	-0.8	-1.8	8.20%
TiTiTiVCl8	8.18%	54.1	-2.6	1.8	-1.7	-4.6	-1.8	-5.4	8.18%
TiTiTiCrCl8	8.17%	52.7	-7.4	-0.1	-7.5	8.7	-1.2	-3.0	8.17%
ScCuCuMgCl8	8.17%	54.1	-62.8	-49.8	-71.0	-35.5	0.7	11.4	5.44%
VTiTiVCl8	8.15%	52.6	-3.5	-0.1	-2.7	8.2	-1.2	-3.5	8.15%
VTiTiCrCl8	8.14%	54.2	-7.1	-1.5	-8.0	4.4	-1.1	-2.5	8.14%

2nd run:

Composition	Fitness	$\Delta H_{60}$	E_decomp_0	E_decomp_1	E_decomp_2	E_decomp_6	$\Delta H_{60}-\Delta H_{61}$	$\Delta H_{60}-\Delta H_{62}$	wH pract.
TiTiCuMgCl8	8.28%	54.9	-34.0	-20.0	-40.2	-19.2	-0.9	5.2	5.52%
ScTiTiTiCl8	8.24%	53.0	0.9	0.1	-1.4	-0.2	-0.7	-5.2	8.24%
TiTiTiTiCl8	8.21%	53.0	0.0	0.0	0.0	0.0	-0.8	-5.7	8.21%
ScTiTiVCl8	8.21%	54.1	-2.4	-1.6	-5.8	-5.0	-1.1	-4.4	8.21%
ScTiTiCrCl8	8.20%	52.6	-8.5	-3.4	-13.8	7.7	-0.8	-1.8	8.20%
TiTiVTiCl8	8.18%	54.1	-2.6	1.8	-1.7	-4.6	-1.8	-5.4	8.18%
TiTiTiCrCl8	8.17%	52.7	-7.4	-0.1	-7.5	8.7	-1.2	-3.0	8.17%
ScCuCuMgCl8	8.17%	54.1	-62.8	-49.8	-71.0	-35.5	0.7	11.4	5.44%
TiVVTiCl8	8.15%	52.6	-3.5	-0.1	-2.7	8.2	-1.2	-3.5	8.15%
TiVTiCrCl8	8.14%	54.2	-7.1	-1.5	-8.0	4.4	-1.1	-2.5	8.14%

3rd run:

Composition	Fitness	$\Delta H_{60}$	E_decomp_0	E_decomp_1	E_decomp_2	E_decomp_6	$\Delta H_{60}-\Delta H_{61}$	$\Delta H_{60}-\Delta H_{62}$	wH pract.
CaTiTiScCl8	8.31%	54.8	22.0	14.1	11.1	25.3	0.9	-3.9	8.31%
CuTiTiMgCl8	8.28%	54.9	-34.0	-20.0	-40.2	-19.2	-0.9	5.2	5.52%
ScNiCaCaCl8	8.28%	52.6	-10.4	-8.8	-20.6	34.3	0.7	2.3	5.52%
TiTiTiScCl8	8.24%	53.0	0.9	0.1	-1.4	-0.2	-0.7	-5.2	8.24%
CaTiCuCaCl8	8.21%	53.7	-8.3	2.3	-21.2	27.0	-1.5	2.0	8.21%
TiTiVScCl8	8.21%	54.1	-2.4	-1.6	-5.8	-5.0	-1.1	-4.4	8.21%
CrTiTiScCl8	8.20%	52.6	-8.5	-3.4	-13.8	7.7	-0.8	-1.8	8.20%
VTiTiTiCl8	8.18%	54.1	-2.6	1.8	-1.7	-4.6	-1.8	-5.4	8.18%
CrTiTiTiCl8	8.17%	52.7	-7.4	-0.1	-7.5	8.7	-1.2	-3.0	8.17%
ScCuCuMgCl8	8.17%	54.1	-62.8	-49.8	-71.0	-35.5	0.7	11.4	5.44%

Table S2: Details on top ten candidates from the three runs. Units are in kJ/mol per formula unit containing one metal atom for decomposition energies, and kJ/(mol·NH<sub>3</sub>) for ΔHs. ΔXY signifies the reaction from an ammine phase with X ammonia molecules to a phase with Y ammonia molecules and X-Y free ammonia gas molecules. The stabilities (E\_decomp\_X) of the mono- and di ammine mixtures are also included, because it is highly relevant to check whether the intermediate ammines will decompose if they are formed, and thereby separating the material and making the desorption irreversible. ΔH<sub>60</sub>-ΔH<sub>6X</sub> determines if intermediate ammine phases are expected to be observed, if it is, wH practical is lowered, for details see the main text.